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MAGNETIC PROPERTIES OF THE TERNARY COMPOUNDS CeT_2Si_2 AND UT_2Si_2

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We have investigated the magnetic properties of the intermetallic compounds CeT_2Si_2 and UT_2Si_2, with T a transition metal. From our measurements we have determined a trend from Pauli-paramagnetism via antiferromagnetism to canted antiferromagnetism with increasing number of d-electrons. Superconductivity, like in $CeCu_2Si_2$, may be found near the paramagnetism/antiferromagnetism boundary.

The ternary compounds MT_2X_2 with M = RE, Th, U and T = 3d-, 4d- 5d-transition metal and X-Si, Ge, have attracted much interest, because of the great variety in their magnetic and superconducting properties. In this report we focus upon the compounds CeT_2Si_2 and UT_2Si_2, because of the ambivalent character of the f-ions. Ce- and U-atoms can either be magnetic or nonmagnetic and this ambivalence can lead to an interplay between magnetism and superconductivity. It was shown that $CeCu_2Si_2$ becomes superconducting below 0.5 K, in spite of the occurrence of local Ce-moments at higher temperatures [1]. More recently, superconductivity was also found in URu_2Si_2, which even shows a magnetic transition at 17 K [2]. We have studied the magnetic properties of the CeT_2Si_2 and UT_2Si_2 compounds systematically, in order to survey the magnetic properties, and to indicate where superconductivity in these systems may be expected.

The polycrystalline samples were prepared by arc melting the pure element in a stoichiometric ratio in a Ti gettered an argon atmosphere. The samples were characterized by X-ray and microprobe analysis. All X-ray powder diffractograms were indexed on basis of the body centered tetragonal $ThCr_2Si_2$-type structure. The resulting lattice parameters are given in tables 1 and 2. This crystal structure has the Bragg-reflection condition that the sum of the Miller indices must be even. This condition was fulfilled for all compounds except for $CePt_2Si_2$ and UT_2Si_2 with T = Ir, Pt and Au. Here, additional lines were observed that could be indexed with an odd sum of Miller indices, which means that these compounds either adopt the primitive tetragonal $CaBe_2Ge_2$-type crystal structure or that the T and Si atoms randomly occupy both the 4(d) and 4(e) sites. However, powder diffractograms cannot distinguish these two possibilities. Also it is not clear whether these crystal structures occur as a low and high temperature modification [3], and this will be a subject for our further investigation.

The magnetic measurements were performed using a Foner vibrating sample magnetometer from 1.5 K to 300 K in magnetic fields up to 5 T. Additionally the ac-susceptibility of all samples was measured in a ^{3}He-cryostat to 0.33 K, and for some selected compounds in a dilution refrigerator down to 30 mK.

It is known that the behavior of Ce-compounds can be dominated by valency fluctuations between Ce^{3+} and Ce^{4+} with corresponding moment fluctuations between 2.54 and $0.0\mu_B$/Ce. For T = Ni we found a Pauli-paramagnetic behavior, and for T = Cu we found the well known Kondo-lattice behavior, with a Ce-moment at higher temperature and superconductivity at 0.59 K. When T is a 4d-metal we again find a Kondo-lattice system for T = Ru, similar to $CeCu_2Si_2$ [4]. For T = Rh, Pd, Ag, Pt Au an antiferromagnetic ordering takes place, with a rearrangement for T = Rh at 5 K and a canting for T = Ag at 4 K.

The magnetism of the U-compounds is in some respects similar to that of the Ce-compounds. However, the 4f-electrons are very localized, whereas the 5f-electrons are more itinerant. It is generally believed that this leads to magnetic moments on the U-atoms if the U–U separation distance is too large for overlap of the 5f-wave functions, and to Pauli-paramagnetism at small U–U-separation. This criterion was formulated by Hill, with a critical separation of about 3.5 Å [5]. In the compounds presently under investigation the U–U separation equals the lattice parameter a, and is always larger than the Hill-limit, independent of the T-metal. Yet, we find here both Pauli-paramagnetc and antiferromagnetic systems and there is no correlation between the U–U separation and the magnetic ordering temperature. Thus the Hill-criterion is violated.

For the UT_2Si_2-compounds, with T a 4d-metal, we found for T = Ru, Rh, Pd an antiferromagnetic ordering. For T = Rh we also found a small ferromagnetic component, and for T = Pd an even larger one. When T is a 5d-metal we observed Pauli-paramagnetism for T = Re, Os and antiferromagnetism for T = Ir, Pt, Au with a ferromagnetic canting for T = Au (see fig. 1).

From tables 1 and 2 we can detect several similarities in magnetic behavior, if we compare the different series of compounds (a series is defined with T either a 3d-, 4d- or 5d-metal). First, we see that the effective moment is almost constant within a series. Second, we observe an "increase" of magnetism within a series from Pauli-

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Table 1
Structural and magnetic parameters of the CeT_2Si_2 compounds. a and c are the lattice parameters, T_N the magnetic ordering temperature, θ_{CW} the Curie–Weiss temperature and μ_{eff} the effective moment per formula unit. pp denotes Pauli-paramagnetism, Ko and Kondo-lattice system, sc superconductivity and (ca) af (canted) antiferromagnetism

T = 3d-		Mn	Fe	Co	Ni	Cu
				pp	pp	Ko + sc
a	(Å)			3.953	4.036	4.105
c	(Å)			9.776	9.575	9.934
T = 4d-		Tc	Ru	Rh	Pd	Ag
			Ko	af	af	ca af
a	(Å)			4.098	4.230	4.250
c	(Å)			10.19	9.873	10.66
T_N	(K)			37.	10.5	9.5
θ_{CW}	(K)			−163.	−57.	−36.
μ_{eff}	μ_B			2.43	2.55	2.54
T = 5d-		Re	Os	Ir	Pt	Au
					af	af
a	(Å)				4.253	4.310
c	(Å)				9.798	10.20
T_N	(K)				6.	10.1
θ_{CW}	(K)				−85.	−18.
μ_{eff}	μ_B				2.42	2.43

Table 2
Structural and magnetic parameters of the UT_2Si_2 compounds. The parameters are defined as in table 1

T = 4d-		Tc	Ru	Rh	Pd	Ag
			Ko + sc	af	af	
a	(Å)		4.127	4.012	4.121	
c	(Å)		9.610	10.06	10.19	
T_N	(K)		17.	130.	97.	
θ_{CW}	(K)		−160.	−40.	−10.	
μ_{eff}	μ_B		2.86	2.65	2.88	
T = 5d-		Re	Os	Ir	Pt	Au
		pp	pp	af	af	ca af
a	(Å)		4.121	4.088	4.217	4.228
c	(Å)		9.681	9.790	9.704	10.26
T_N	(K)			5.5	36.	78.
θ_{CW}	(K)			−156.	−57.	−36.
μ_{eff}	μ_B			3.03	3.22	3.11

paramagnetism via antiferromagnetism to canted antiferromagnetism with increasing number of d-electrons. Finally, we note a decrease of the Curie–Weiss temperatures with decreasing number of d-electrons within a series. This leads to absolute values up to 25 times the

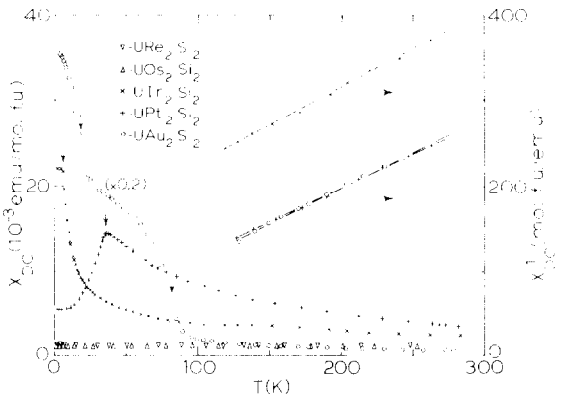

Fig. 1. DC susceptibility and inverse susceptibility of the UT_2Si_2 compounds with T a 5d-metal, measured in a field of 1 T.

Néel temperature at the Pauli-paramagnetic antiferromagnetic phase boundary.

We conclude from these observations that the magnetism is not determined by the Ce–Ce or U–U distance, but by the number of d-electrons of the T-metal. With decreasing number of d-electrons the magnetic moments will be more and more compensated by the conduction electrons, leading to more negative values of the Curie–Weiss intercept temperatures and to a transition from antiferromagnetism to Pauli-paramagnetism. The most interesting behavior may be found at the borderline cases between Pauli-paramagnetism and antiferromagnetism, where a Kondo-lattice may arise as for $CeCu_2Si_2$. Similar arguments holds for URu_2Si_2. These two compounds are superconductors. Therefore, we suggest that superconductivity might also be found in other "borderline" compounds like $CeRu_2Si_2$ and UIr_2Si_2. This possibility has already been checked for $CeRu_2Si_2$, but no superconductivity was found down to 40 mK [4]. Also pseudo-ternary compounds like $U(Os,Ir)_2Si_2$ might be very interesting in this respect.

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